

LA-UR- 99 - 2201

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Title: NON-THERMAL PLASMA TECHNIQUES FOR AIR TREATMENT

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Submitted to: 5th International Conference on Advanced Oxidation Technologies for Water & Air Remediation (AOTs-5)
Albuquerque, NM; May 23-28. 1999
Workshop on Advanced Oxidation Technologies:
Fundamentals & Applications

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Form 836 (10/96)

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REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 074-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing this collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (Leave blank)			2. REPORT DATE 1999	3. REPORT TYPE AND DATES COVERED Conference Proceedings, May 23-28, 1999
4. TITLE AND SUBTITLE Non-Thermal Plasma Techniques for Air Treatment			5. FUNDING NUMBERS N/A	
6. AUTHOR(S) Louis A. Rosocha				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Los Alamos National Laboratory			8. PERFORMING ORGANIZATION REPORT NUMBER N/A	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) SERDP 901 North Stuart St. Suite 303 Arlington, VA 22203			10. SPONSORING / MONITORING AGENCY REPORT NUMBER N/A	
11. SUPPLEMENTARY NOTES No copyright is asserted in the United States under Title 17, U.S. code. The U.S. Government has a royalty-free license to exercise all rights under the copyright claimed herein for Government purposes. All other rights are reserved by the copyright owner.				
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13. ABSTRACT (Maximum 200 Words) To prevent toxic substances from entering and spreading through the environment and to meet increasingly stringent regulations, more effective methods of destroying chemical pollutants are being examined. Advanced oxidation processes, historically used for the treatment of recalcitrant water pollutants via hydroxyl radicals (OH), are now being applied to the treatment of gas-phase pollutants using non-thermal plasmas (NTPs). In NTPs, chemical decomposition can also be initiated by radicals other than OH, some of which may even be reductive rather than oxidative. Non-thermal or non-equilibrium plasmas in gaseous media are characterized by combined energetic-electron (e.g., 1-1- eV) and low-temperature gas conditions. Free radicals, particularly oxygen-atom radicals, are effectively generated by NTPs and play a key role in the process of ozone generation.				
14. SUBJECT TERMS SERDP, non-thermal plasma technique, pollutant, oxidation				15. NUMBER OF PAGES 43
				16. PRICE CODE N/A
17. SECURITY CLASSIFICATION OF REPORT unclass	18. SECURITY CLASSIFICATION OF THIS PAGE unclass	19. SECURITY CLASSIFICATION OF ABSTRACT unclass	20. LIMITATION OF ABSTRACT UL	

Non-Thermal Plasma Techniques for Air Treatment

AOTs-5 Workshop Advanced Oxidation Technologies: Fundamentals & Applications

Louis A. Rosocha

Los Alamos National Laboratory



May 24, 1999 - Albuquerque, NM

LA-UR-99-2201 (Unclassified, Distribution Unlimited)

Non-Thermal Plasma Techniques for Air Treatment

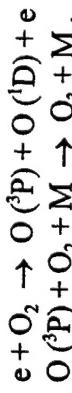
AOTs-5 Workshop on Advanced Oxidation Technologies: Fundamentals & Applications
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Louis A. Rosocha
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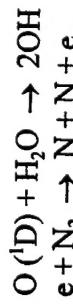
Abstract

To prevent toxic substances from entering and spreading through the environment and to meet increasingly stringent regulations, more effective methods of destroying chemical pollutants are being examined. Advanced oxidation processes, historically used for the treatment of recalcitrant water pollutants via hydroxyl radicals (OH), are now being applied to the treatment of gas-phase pollutants using non-thermal plasmas (NTPs). In NTPs, chemical decomposition can also be initiated by radicals other than OH, some of which may even be reductive rather than oxidative.

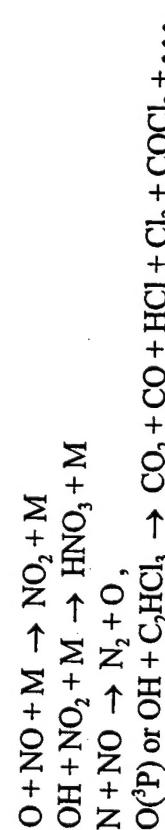
Non-thermal or non-equilibrium plasmas in gaseous media are characterized by combined energetic-electron (e.g., 1-10 eV) and low-temperature gas conditions. Free radicals, particularly oxygen-atom radicals, are effectively generated by NTPs and play a key role in the process of ozone generation (the historical source of OH for water treatment):



Other free radicals like the hydroxyl radical and nitrogen-atom radicals are also generated with NTPs in gases:



In the past several years, there has been increased interest in innovative pollution-control techniques. Researchers have explored the treatment of gas-phase pollutants with AOPs. Electric-discharge and electron-beam generated non-thermal plasmas are considered a promising technology for some applications, e.g., the removal of oxides of nitrogen and sulfur (NO_x and SO_x) and hydro/chloro-carbon volatile organic compounds (VOCs) from air-emissions streams:



In this talk, we will discuss non-thermal plasmas in the context of an AOP - namely a tool for free-radical-initiated decomposition reactions. The main points covered are the motivation for and applications of the process, background of atmospheric-pressure, non-thermal plasma processing for air-pollution control, figures-of-merit and cost examples, and representative application examples of the technology.

Thanks to

Prof. J.S. Chang & Dr. Kuniko Urashima. McMaster University

Dr. S.J. Kim, Korean Institute of Machinery and Materials

Dr. Andrzej Miziolek, Army Research Laboratory

and

the US Strategic Environmental Research & Development
Program (SERDP)

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Outline of talk

- Motivation for and applications of research
- Background of atmospheric-pressure, non-thermal plasma processing for environmental applications (air-pollution control)
 - Basics of radical formation & decomposition chemistry
 - Figures-of-merit & costs for decomposition of example pollutants
- Representative application examples: environmental remediation of VOCs and removal of NO_x from engine test facilities
- Future trends
- Summary

Technology-development motivators are regulatory, economic, and social

Regulatory

- New and/or more stringent regulations
- Clean Air Act Amendment of 1990, Clean Water Act
- Attendant MACT standards

- Enforcement of regulations

Economic

- Impact of regulations
- Costs associated with conventional methods

Social

- Environmental consciousness
- Health risks

The CAAA 1990 contains three titles closely related to new technologies (e.g., NTPs)

Title III (Air Toxics)

- Directly concerned with emission of hazardous air pollutants (HAPs)
- Established standards for 189 listed chemicals

Title IV (Acid Rain)

- Reduction of SOx/NOx emissions

Title VI (Stratospheric Ozone)

- Phase out of VOCs

MACT (Max Achievable Control Technology) Standards

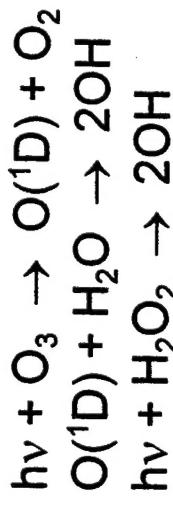
- Calls for state-of-art pollution control equipment
- Require changes in equipment, processes, treatment

Non-thermal plasmas (NTPs) are a type of advanced oxidation & reduction process which use free radicals to decompose pollutants in the gas phase.

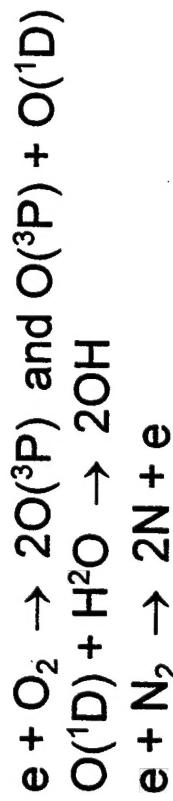
Non-thermal plasmas are a type of AOP making use of “cold combustion” via free-radical reactions.	Applications <ul style="list-style-type: none">Flue gases: e.g., NO_x & SO_xVOCS: e.g., hydrocarbons & halocarbonsOdors: H_2S, others
The key idea is to direct electrical energy into favorable chemistry for oxidizing and/or reducing pollutants to more manageable forms (simpler or mineralized terminal products).	Potential Advantages <ul style="list-style-type: none">In-situ generation of chemical reactantsNo added fuel (greenhouse gases)Simultaneous removal of multiple pollutant speciesElectronic feedback for optimal process control.

Historically, Advanced Oxidation Processes (AOPs) have involved ozone (O_3) and hydroxyl-radical (OH) reactions.

OH radicals were originally generated by the photolysis of O_3 and/or H_2O_2 in aqueous solution:*



The field has expanded to gas-phase processes in which both oxidative and reductive radicals are generated by electron-induced dissociation:



Not the whole story - O_3 & H_2O_2 can directly generate OH in water.

Prospective applications of NTP technology to off-gas treatment:

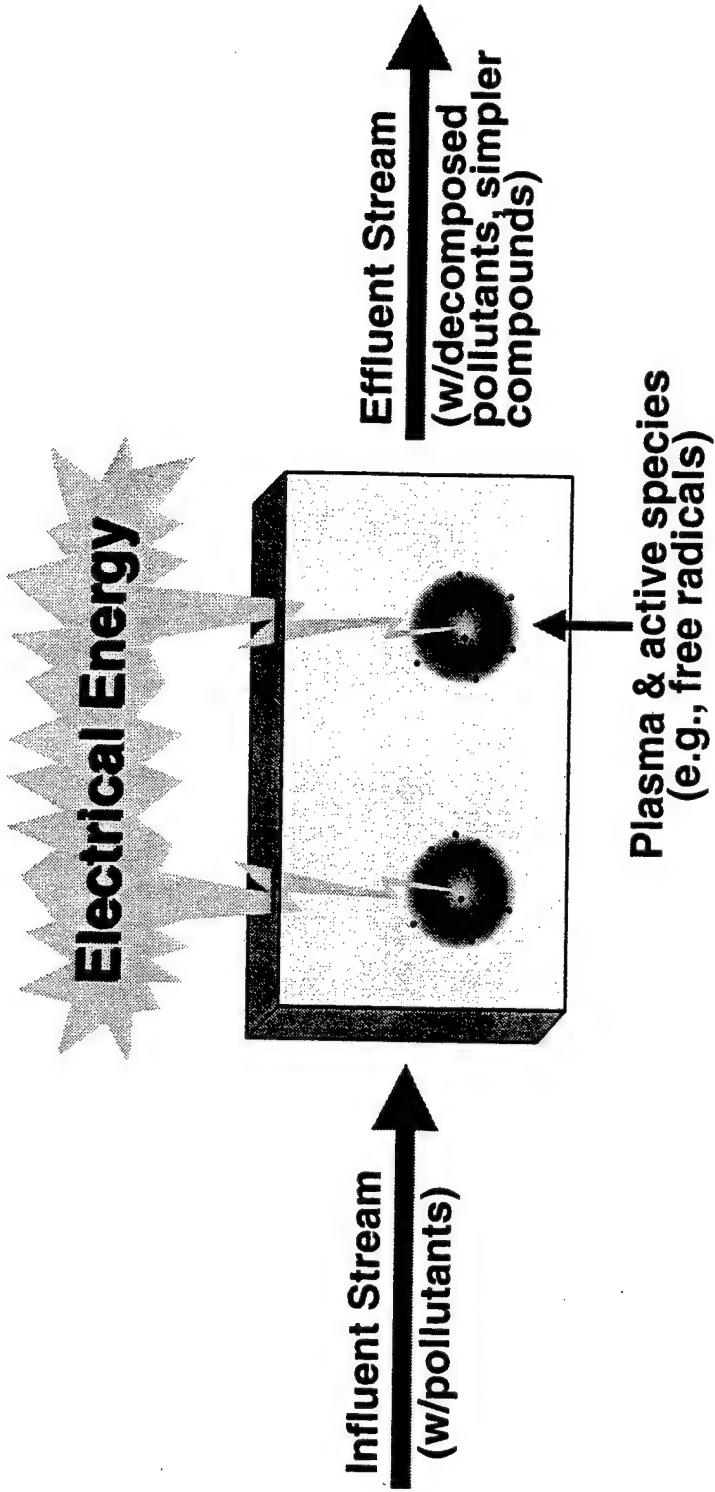
Direct processing (single-stage equipment)

- Remediation of solvents extracted from soil or groundwater
- Processing of industrial stack-gases or flue-gases
- Control of air emissions from waste processing (e.g., incinerators)
- Control of air emissions from diesel motor-generators, jet engine test facilities

Indirect processing (staged equipment)

- Destruction of hazardous organic liquids
- Treatment of heterogeneous wastes
- Processing of special materials

Non-thermal Plasmas Decompose Pollutants Via Active Species Generated in the Process Gas



The radical production efficiency (G-value) depends on the gaseous electronics. Radical generation is mainly initiated by energetic-electron collisions.

$$G = f \left(\frac{k_{rad}}{V_d \frac{E}{N}} \right)$$

- E/N is the reduced field,
- V_d is the electron drift velocity, which depends on E/N ,
- k_{rad} is the rate constant for radical formation (e.g., a dissociation rate constant, which depends on E/N), and/or other rate constants.

Gas-phase plasma chemical decomposition is driven by electron impact and radical attack.

Plasma chemical decomposition of VOCs produces a variety of terminal products.

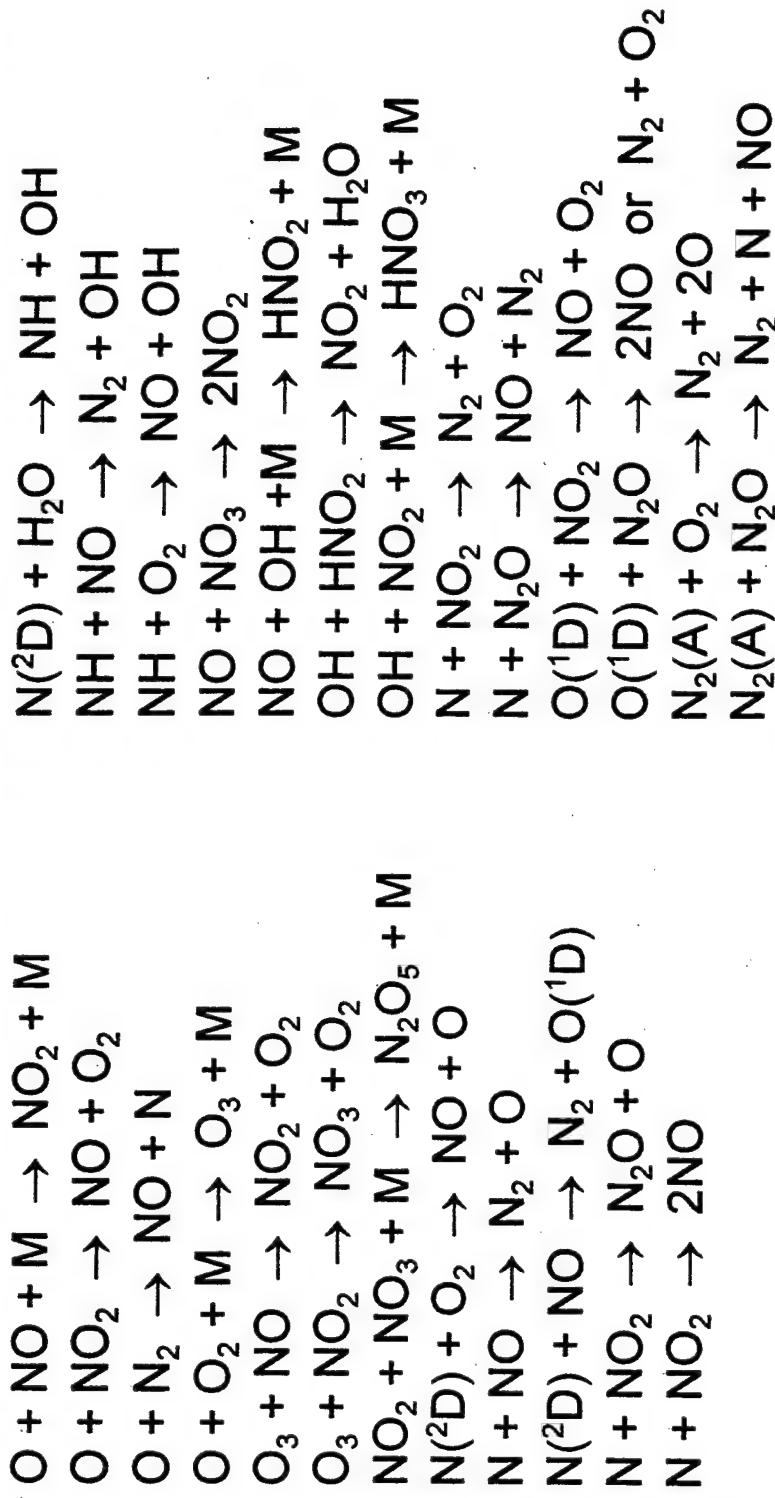
(1) $e + X \rightarrow \text{products}$
(2) $O, OH, N, \text{etc.} + X \rightarrow \text{products}$

- The first reaction is dominant at large pollutant mole fractions.
- The second dominates at smaller mole fractions.
(More energy is directly absorbed by pollutant at high mole fraction, hence electron channel dominates.)

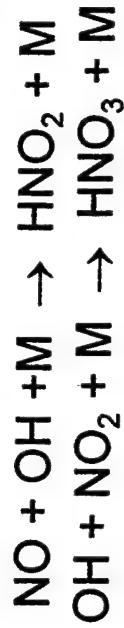
- $e, O, OH, N, \text{etc.} + X \rightarrow \text{products}$**
- Manageable products: $CO_2, CO, Cl_2, HCl, COCl_2$
- Undesirable products: Other halocarbons, hazardous byproducts (e.g., DCAC - $CHCl_2COCl$); polymers

- Secondary treatment:
 $CO_2 + NaOH \rightarrow NaHCO_3$
 $CO + \text{catalyst} \rightarrow CO_2$
 $Cl_2 + NaHCO_3 \rightarrow NaCl + HCl + \dots$
 $HCl + NaOH \rightarrow NaCl + H_2O$
 $COCl_2 + H_2O \rightarrow 2HCl + CO_2$

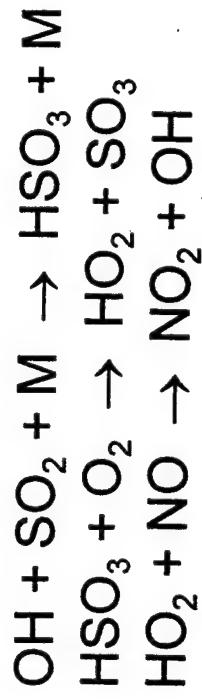
Major de- NO_x Reactions in Moist Gas Mixtures w/o HCs



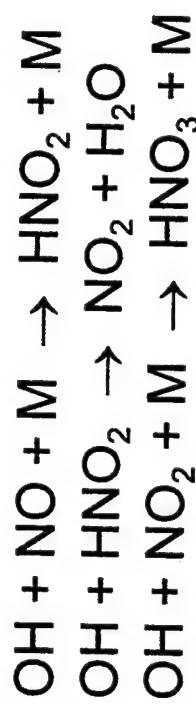
Primary Acid-Formation Pathways:



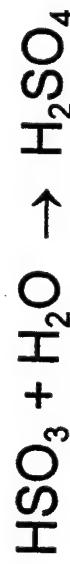
The presence of SO_2 recycles OH radicals & reduces effective de- NO_x energy cost



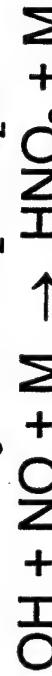
The OH radical then goes on to be used again in de- NO_x



Acid is also formed by the reaction



With ammonia (NH_3) addition, useful particulates (fertilizer) can be formed from NO_x



(Ammonium Nitrate fertilizer)

A generalized removal equation depends on plasma chemistry (radical yields), reaction chemistry, and applied plasma specific energy.

Generalized differential equation:

$$\frac{k[X] + \sum_i k_{S_i} [S_i]}{k[X]} d[X] = -G d\bar{E}$$

Integration with limits $[X]_0 \rightarrow [X]$ and $0 \rightarrow E$ gives:

$$\frac{[X]}{[X]_0} + \frac{\sum_i k_{S_i} [S_i]}{k [X]_0} \ln \frac{[X]}{[X]_0} - 1 = -\frac{G \bar{E}}{[X]_0}$$

For low degree of removal (i.e., $[X]/[X]_0 \sim 1 + \ln \{ [X]/[X]_0 \}$), an analytical solution is obtained:

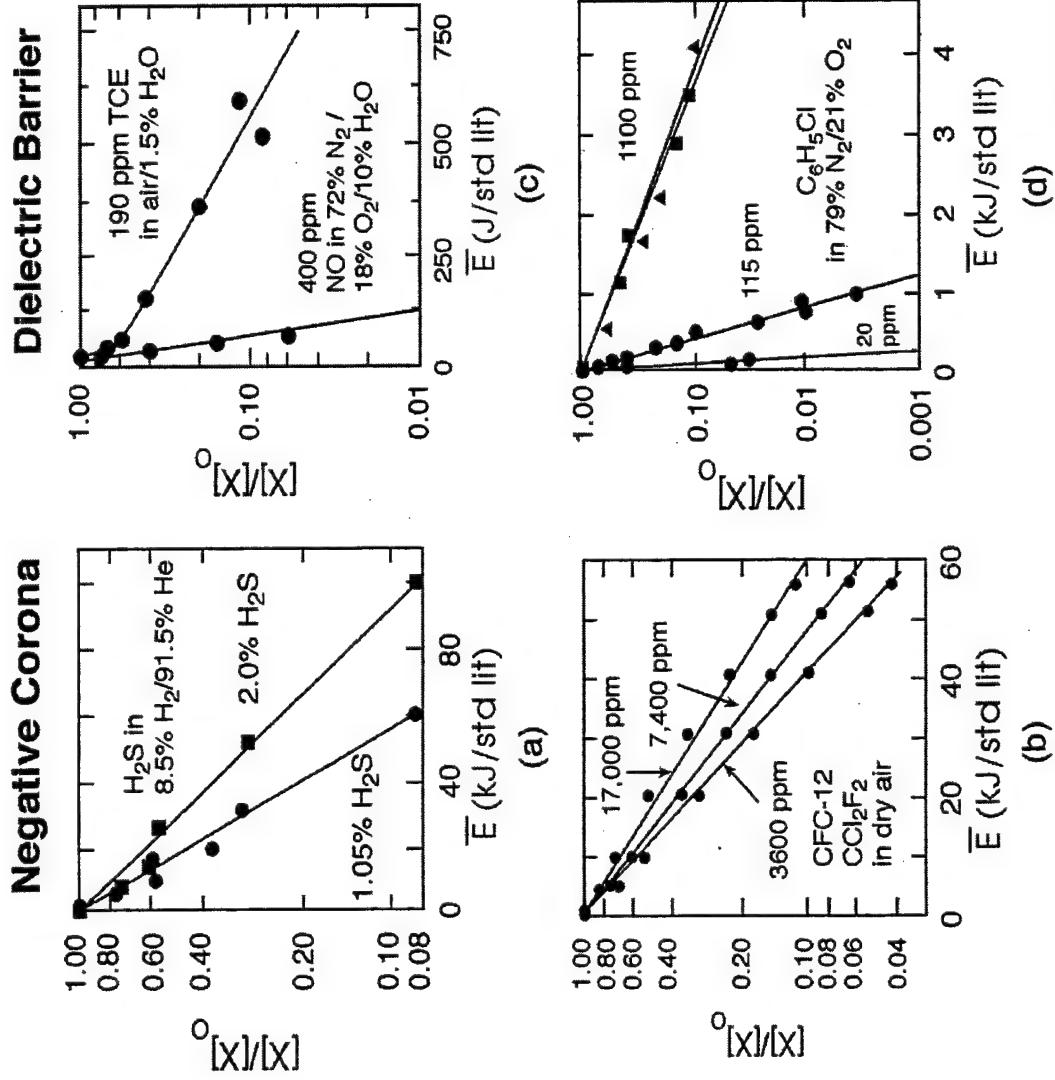
$$[X]/[X]_0 = \exp(-\bar{E}/\beta)$$

where

$$\beta = \frac{1}{G} \left(\sum_i k_{S_i} [S_i] \right)$$

When $k[X] \ll \sum_i k_{S_i} [S_i]$, the β -value and, hence the degree of removal $[X]/[X]_0$ shows no dependence on the initial concentration $[X]_0$.

Example removal data for various compounds



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Scaling laws for pollutant removal

$$(1) \quad [X] = [X]_0 \exp(-\bar{E}/\beta),$$

where $[X]_0$ is the initial pollutant concentration, $[X]$ is the resulting concentration, \bar{E} is the applied specific energy (or plasma power divided by gas flow rate, P/Q), and β is the e-fold energy density. Supplying one β to the reactor reduces the concentration by $1/e$, 2β by $1/e^2$, and so on.

A useful figure of merit for the decomposition of pollutants is defined by the energy delivered to the plasma per hazardous molecule removed from the gas stream. At any instant, this can be expressed as the following quantity obtained by solving Equation 1 for \bar{E} and taking the derivative:

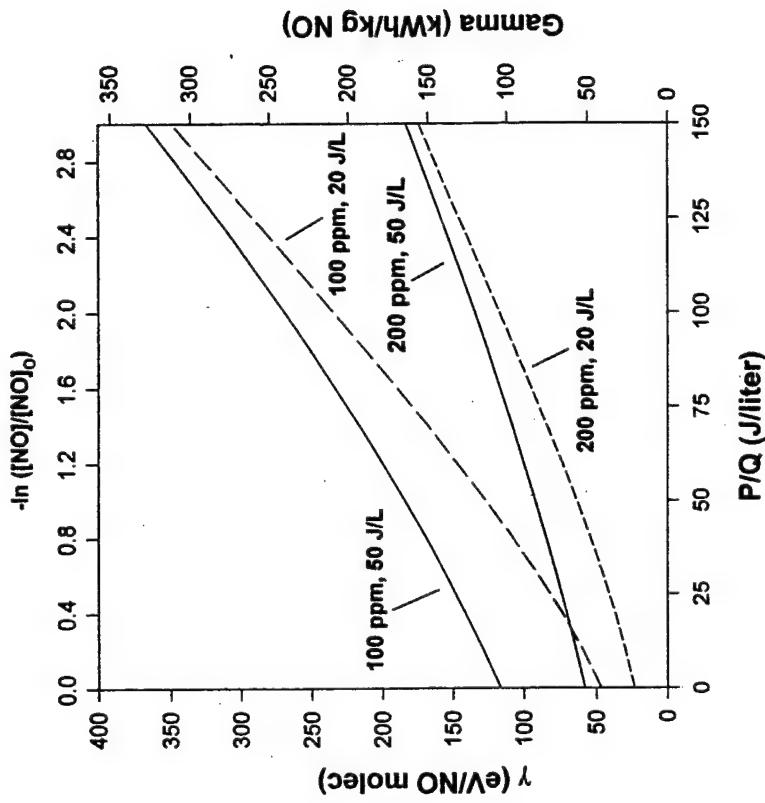
$$(2) \quad \gamma_i = -\frac{d\bar{E}}{d[X]} = -\frac{d[-\beta \ln(\frac{[X]}{[X]_0})]}{d[X]} = \frac{\beta}{[X]}.$$

This is the instantaneous energy cost per molecule removed; a more practically-useful parameter is the integral, or average, energy cost γ

$$(3) \quad \gamma = \frac{\bar{E}}{[X]_0 - [X]} = \frac{-\beta \ln(\frac{[X]}{[X]_0})}{[X]_0 (1 - \frac{[X]}{[X]_0})}.$$

Here, the energy cost per molecule is expressed in terms of the β -value, the degree of removal, and the initial concentration.

Scaling laws for pollutant removal (cont'd)



Energy costs and degree of removal for NO removal in an NO-air mixture with one e-fold plasma specific energies of 50 J/liter and 20 J/liter. NO is one of several compounds whose β -value is not very sensitive to the initial concentration; therefore, the removal energy costs decrease as the concentration increases (note 200 ppm case vs 100 ppm case).

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Example: NO_x - removal power requirement depends on exhaust gas flow rate, characteristic specific energy, and desired degree of removal.

- **Typical removal scaling:** $[\text{X}]/[\text{X}]_0 = \exp(-E_s/\beta)$,

where $E_s = P/Q$, plasma power divided by gas flow rate and β = specific energy for one e-fold removal.

For NO , $\beta \sim 10 \text{ J/lit}$ (e-beam); $\sim 50 \text{ J/lit}$ (electrical discharge).

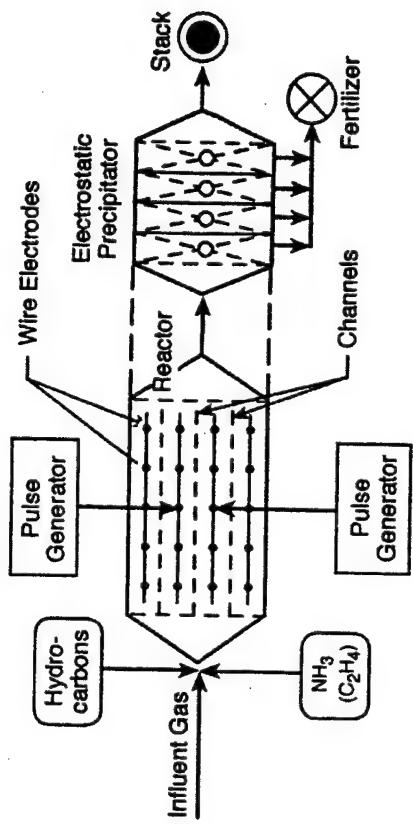
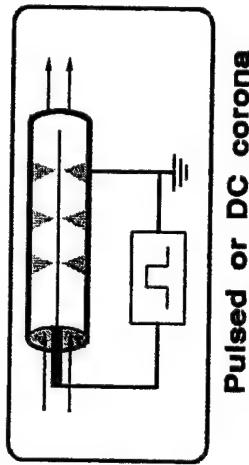
- **Power requirement for one e-fold removal:** $P = Q \cdot \beta$.

Small Source:	Large Source:
$Q \sim 100 \text{ SCFM}$ (2832 lit/min)	$Q \sim 1 \times 10^6 \text{ SCFM}$ ($2.83 \times 10^7 \text{ lit/min}$)
$P \sim 472 \text{ W}$ (e-b), 2.4 kW (e-d)	$P \sim 4.7 \text{ MW}$ (e-b), 24 MW (e-d)

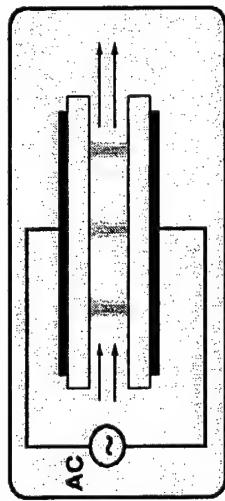
Example NTP-equipment designs

- **Pulsed Corona Plasma Reactor System**
- **Dielectric-Barrier (Silent Discharge) Plasma Reactor System**
- **Electron-Beam Plasma Reactor System**
- **Hybrid/NTP Reactor-Adsorber System: NTP reactor + activated carbon + electrostatic precipitator (ESP)**
- **Plasma-Catalytic System: NTP reactor + catalytic converter.**
- **Corona Radical Shower (CRS) System: Secondary NTP reactor injects radicals formed in bleed gas into main reactor.**

Pulsed Corona System

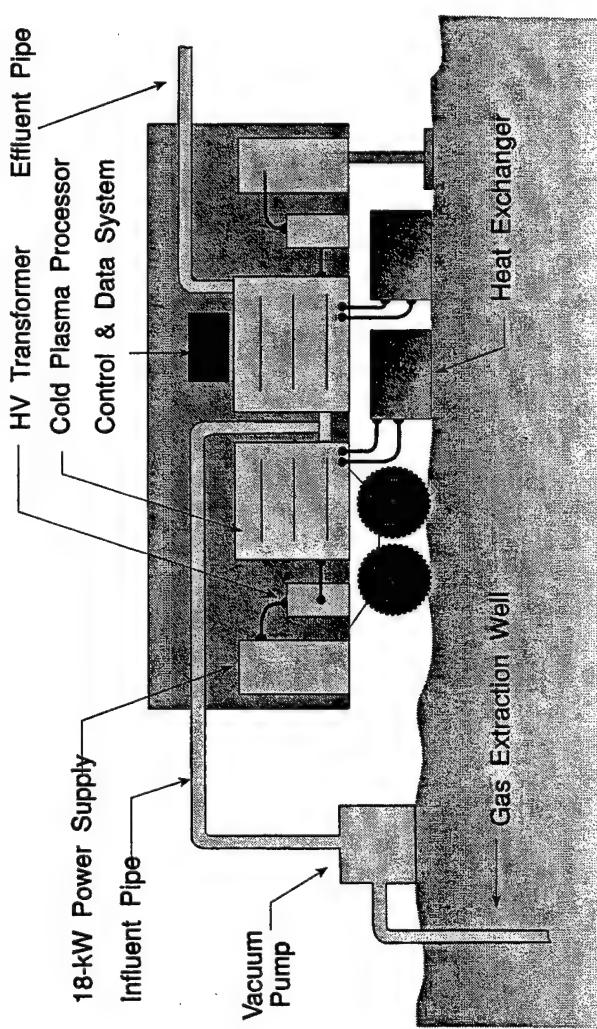


Dielectric-Barrier (Silent Discharge) System



**Silent discharge
(dielectric-barrier discharge)**

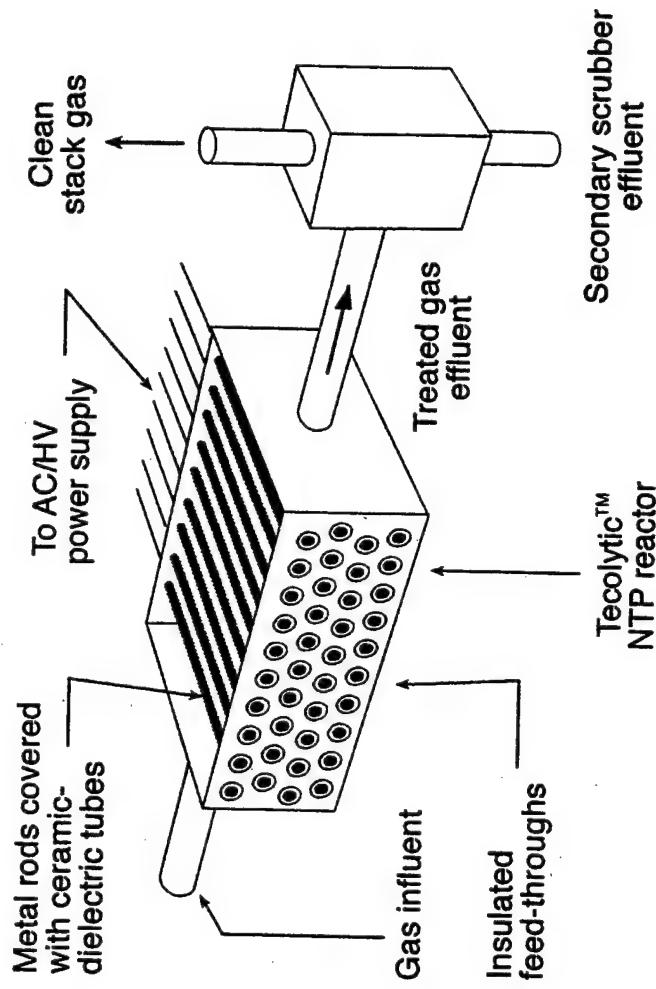
Illustration of mobile dielectric-barrier NTP reactor system employed for VOC decomposition tests at McClellan AFB. Each plasma reactor tank operated at up to 10 kW of plasma power.



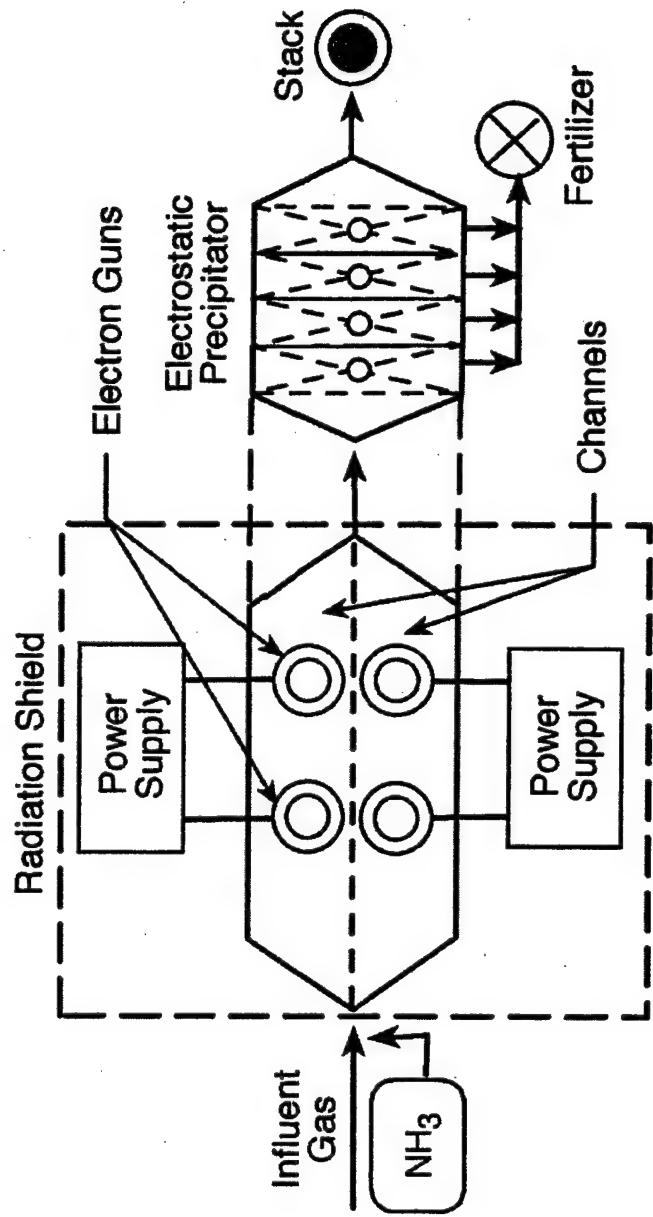
CRADAs with the Electric Power Research Institute (EPRI) & High Mesa Technologies (HMT) were an essential part of the development & fielding of this equipment.

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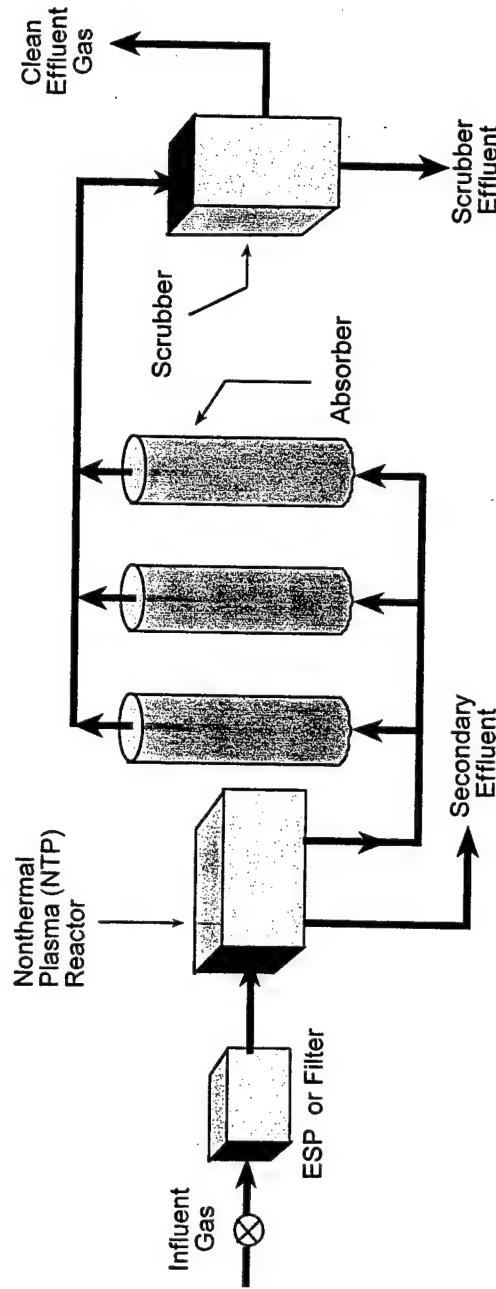
Schematic diagram of commercial Tecolytic™ modified dielectric-barrier NTP reactor system for de- NO_x/SO_x (flue-gas treatment)



Electron-Beam System

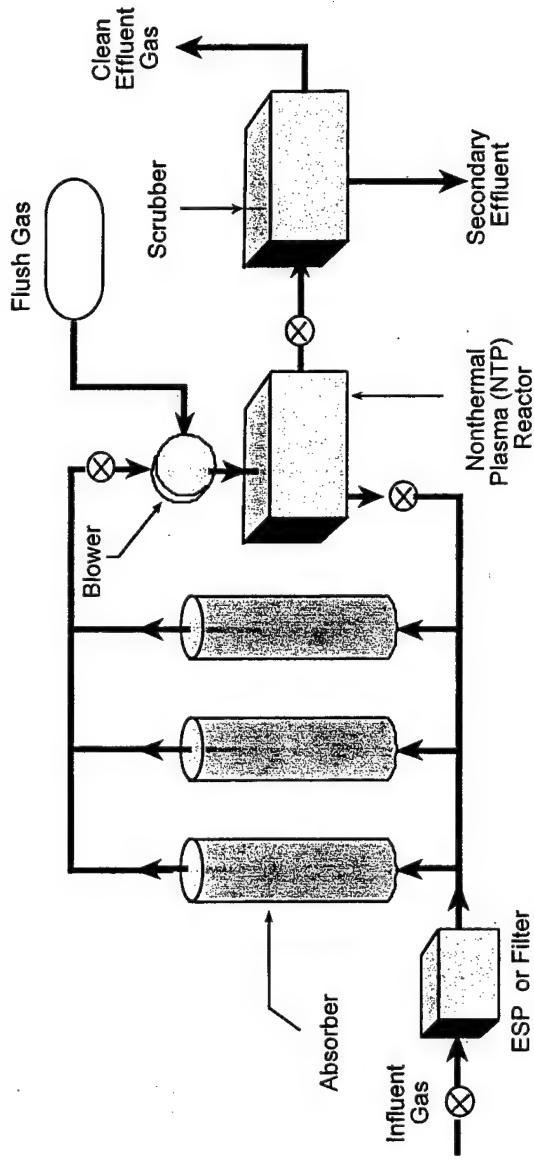


Serial-Mode NTP-Adsorber System



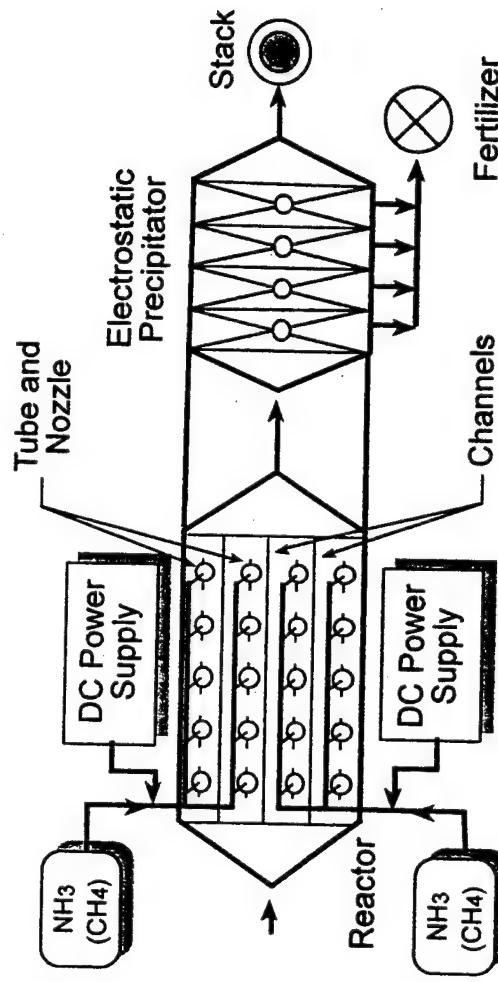
Serial-mode NTP-absorber architecture. Disposal and/or regeneration economic advantage comes from reducing the load on the absorber or converting the pollutants to more easily-absorbed compounds.

Regenerative-Mode NTP-Adsorber System



Regeneration-mode NTP-absorber architecture. Economic and performance advantages may be gained by regenerating the absorbers off-line from pollutant capture, but employing on-site, rather than off-site handling.

Corona Radical Shower System

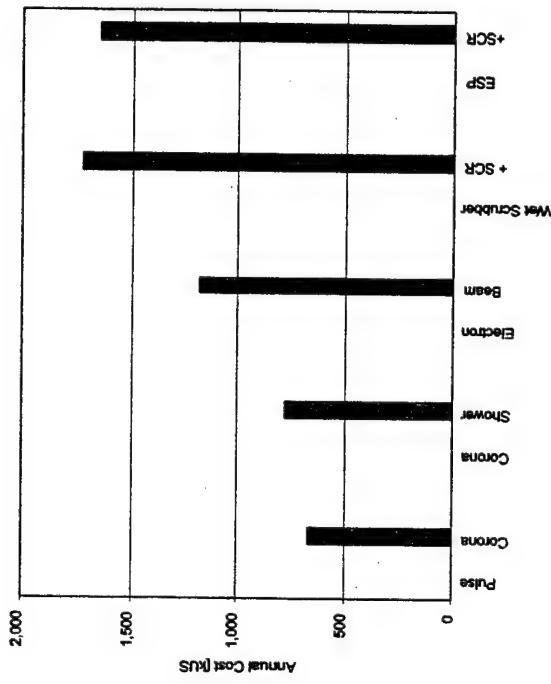


Schematic diagram of CRS reactor. Ammonia (NH_3) or methane (CH_4) are added to generate radicals that drive reactions leading to the formation of particulates; these particulates are then captured by the electrostatic precipitator. Some of the captured products are useful for agricultural fertilizer (e.g., ammonium nitrate, NH_4NO_3).

We have provided economic analyses for three hybrid NTP systems compared to conventional de-NO_x systems

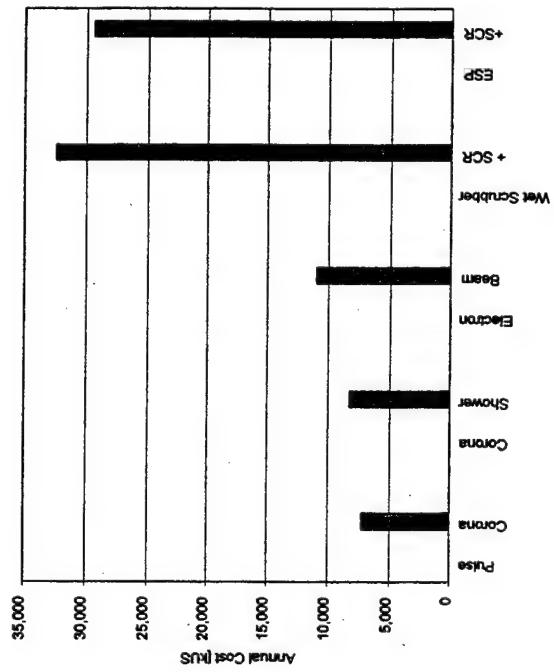
Conventional	NTP
Selective Catalytic Reduction (SCR) + Wet Scrubbers	Pulsed Corona
Selective Catalytic Reduction (SCR) + Electrostatic Precipitator (ESP)	Electron-Beam
	Corona Radical Shower (CRS)

Cost comparisons for various de- NO_x systems



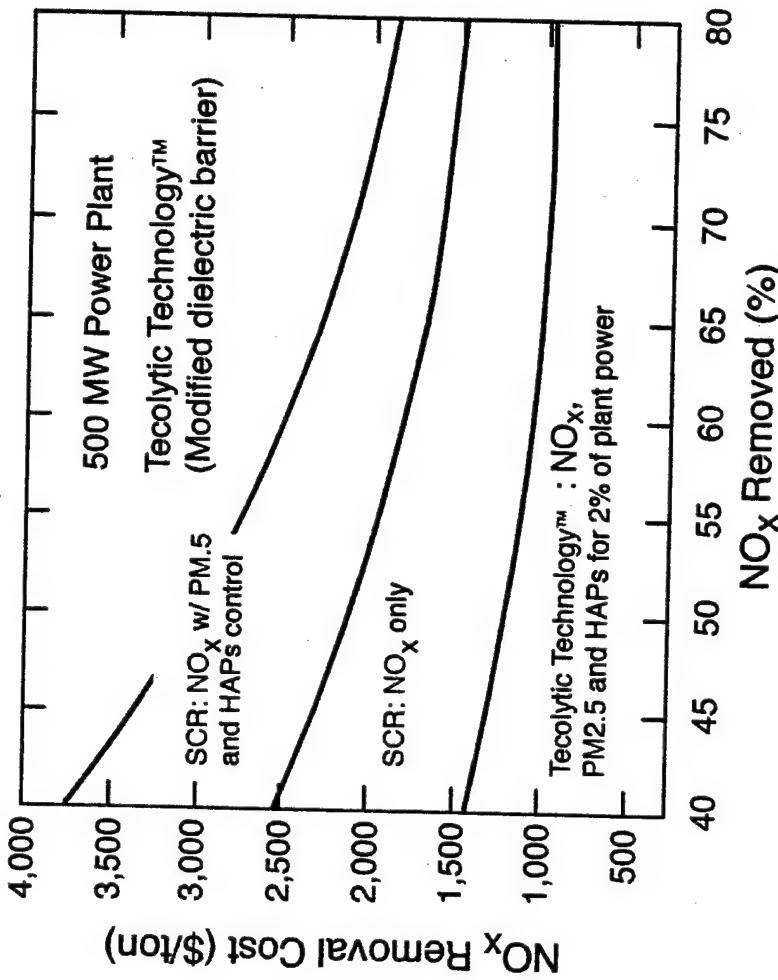
	Gas Flow Rate: $1.0 \times 10^5 \text{ Nm}^3/\text{h}$ ($5.89 \times 10^4 \text{ SCFM}$)			
(k\$)	Pulsed Corona Corona Shower	Electron Beam	Wet Scrubber + SCR	ESP + SCR
Capital Recovery	345	410	718	36
Labor & Maintenance	218	246	377	273
Electric Power	99	116	77	123
Chemicals & Utilities	6	6	6	83
Total Annual Cost	664	774	1,176	1,192
Fertilizer Recovery	4	4	4	0

Cost comparisons for various de-NO_x systems (cont'd.)



Gas Flow Rate: 1.70 x 10 ⁶ Nm ³ /h (1.0 x 10 ⁶ SCFM)				
(k\$)	Pulsed Corona	Corona Shower	Electron Beam	Wet Scrubber + SCR
Capital Recovery	3,594	4,151	6,504	3,802
Labor & Maintenance	1,823	2,060	3,061	4,641
Electric Power	1,674	1,970	1,318	2,082
Chemicals & Utilities	110	110	110	21,935
Total Annual Cost	7,139	8,230	10,931	32,459
Fertilizer Recovery	62	62	62	0

Cost comparisons for commercial Tecolytic™ modified dielectric-barrier NTP reactor system for de- NO_x/SO_x (flue-gas treatment)



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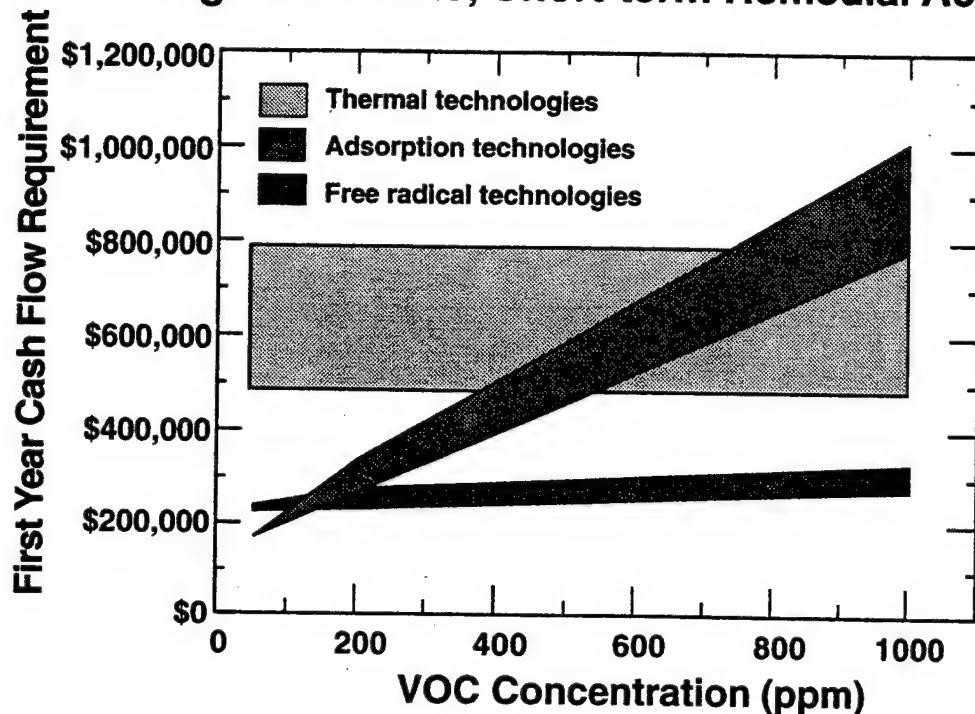
Comparison of SVE off-gas technology cost attributes by general technology type:

Cost Effectiveness Attribute	Technology
Advantageous at low concentrations:	adsorption, free radical
Advantageous at higher concentrations:	thermal, free radical
Advantageous at low flow rates:	adsorption, free radical
Advantageous at higher flow rates:	thermal, free radical
Low capital cost:	adsorption, free radical
Low operational cost:	thermal, free radical
Recycling option:	adsorption
Highest DRE:	thermal, free radical
Performs well at high humidity:	thermal
Easily scalable:	free radical

Free-radical (NTP) technologies show advantages for spanning a broad range of concentrations and gas flow rates in remediation of hydrocarbons (~ 100 J/L β -value). Study from Cummings & Booth, LANL (1996).

Cost Comparisons for Soil-Vapor Extraction (SVE) Off-Gas Treatment Technologies

High Flow Rate, Short-term Remedial Action



High Flow Rate, Long-term Remedial Action

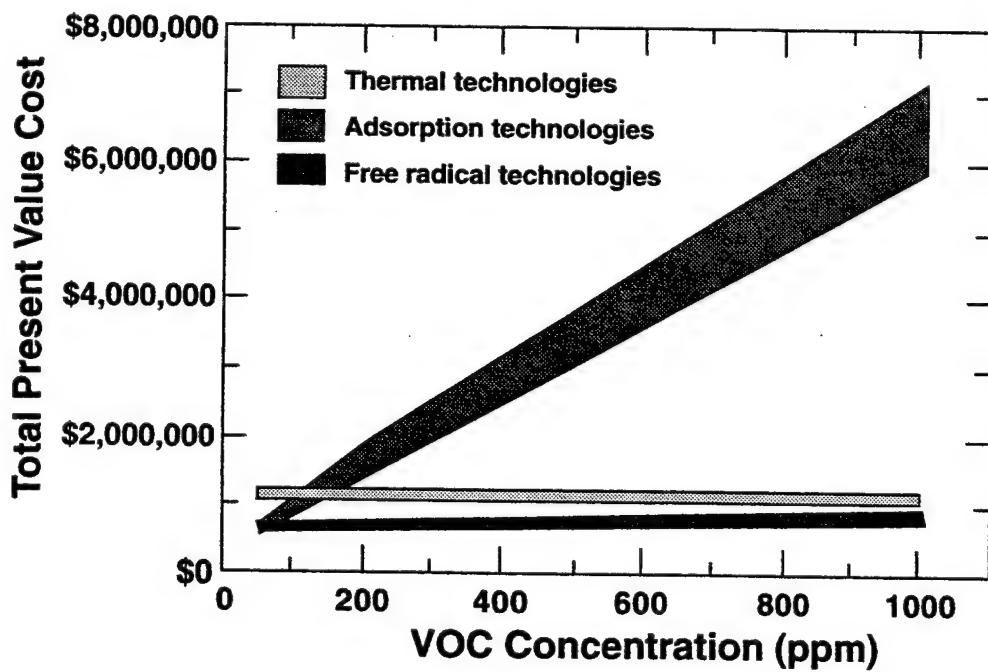


Figure 1. Cost Comparison at Low Flow Rate, Short-term Remedial Action

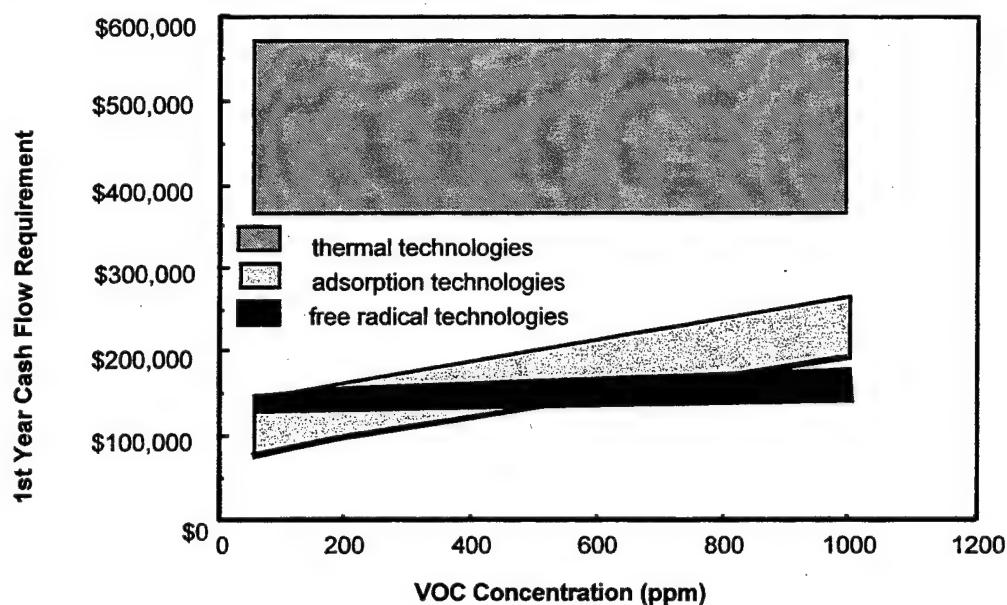
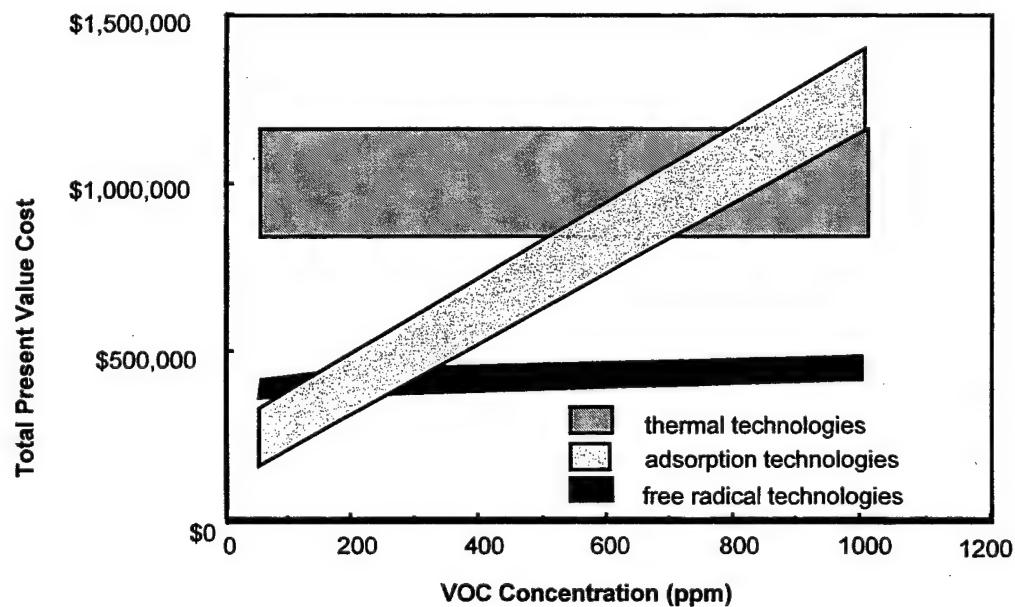


Figure 2. Cost Comparison at Low Flow Rate, Long-term Remedial Action



- **Figure 1** shows adsorption and free radical technologies are competitive at the low flow rate across a range of concentrations, over the short term
- The high capital cost of thermal technologies makes them an impractical choice for remediation under these conditions
- **Figure 2** shows adsorption technologies are cost effective only at low concentrations over the long term
- Free radical technologies are the most cost effective alternatives at concentrations above 200 ppm

Figure 3. Cost Comparison at High Flow Rate, Short-term Remedial Action

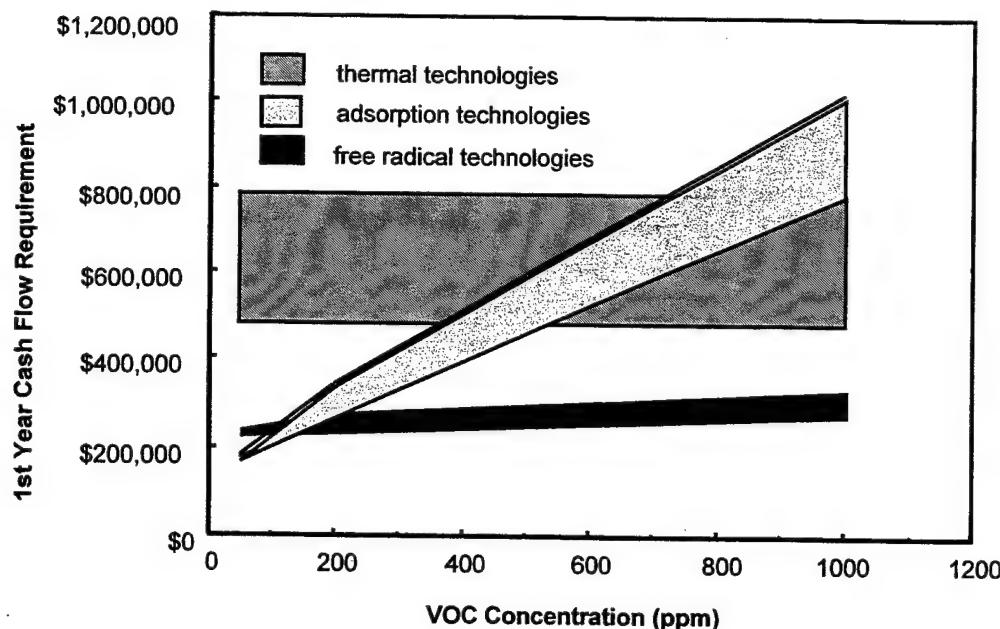
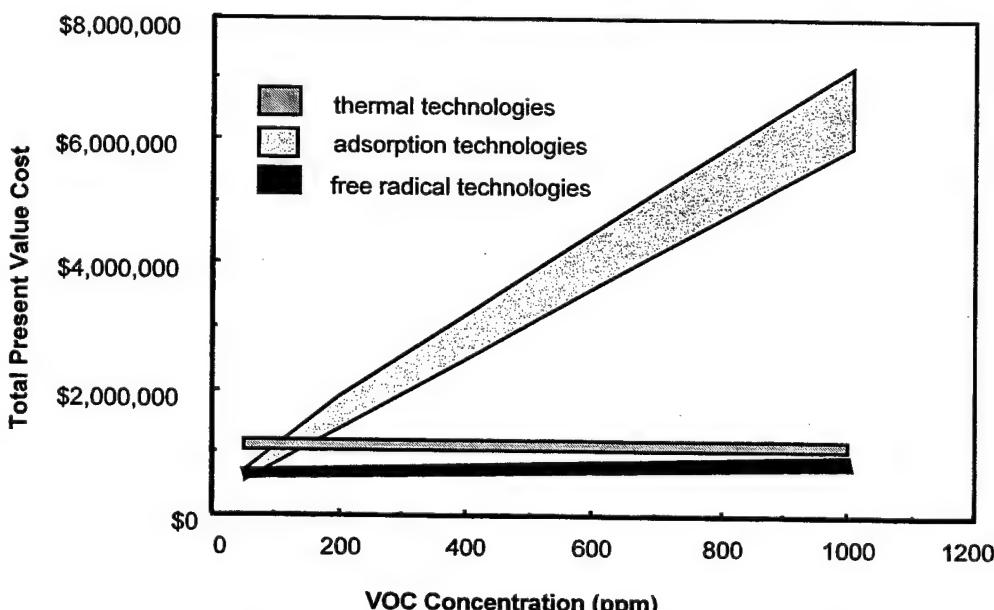
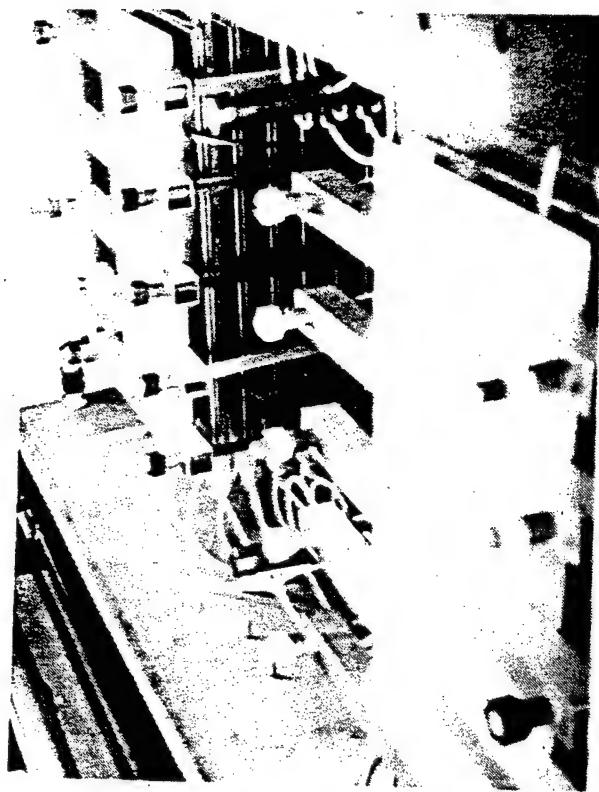


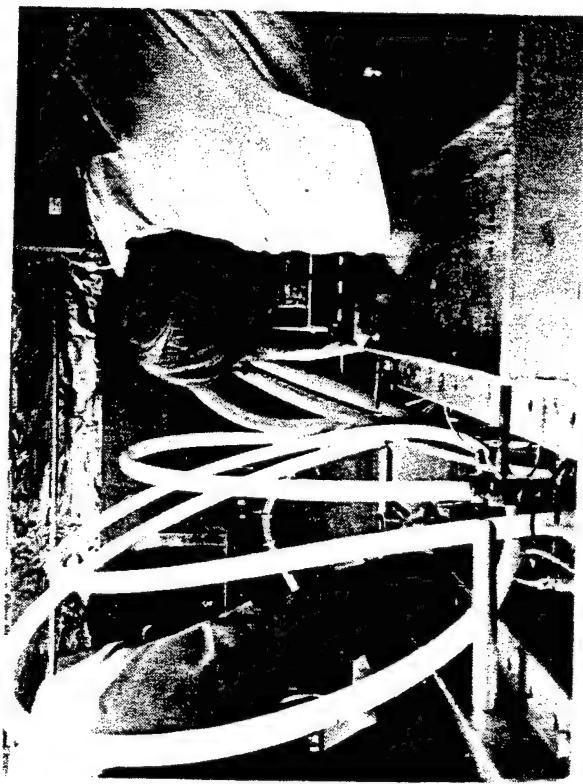
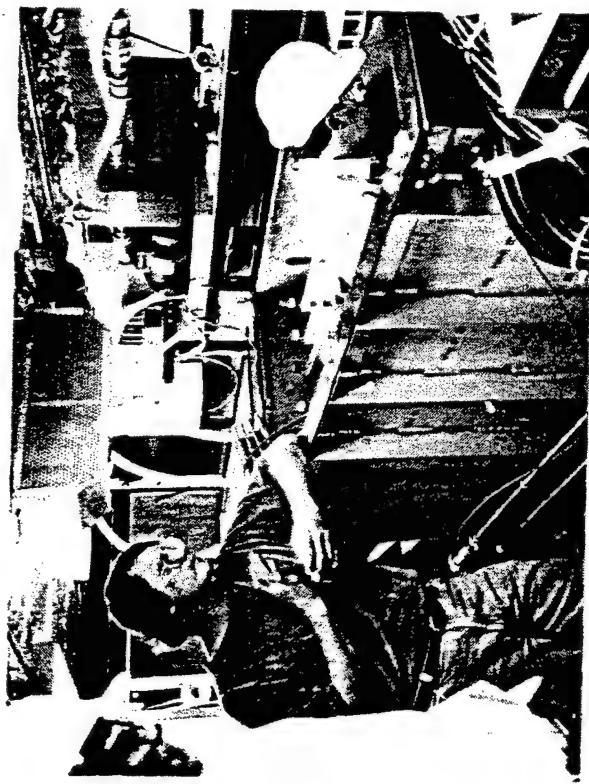
Figure 4. Cost Comparison at High Flow Rate, Long-term Remedial Action



- **Figure 3** shows adsorption and free radical technologies are competitive at the high flow rate only at low VOC concentrations over the short term
- Thermal technologies become competitive with adsorption at higher VOC concentrations
- Free radical technologies are the most cost effective alternatives at concentrations above 200 ppm
- **Figure 4** shows thermal and free radical technologies are the most cost effective choices at the higher flow rate over the long term
- The high operational costs associated with adsorption technologies causes them to lose cost effectiveness over the long-term schedule as concentrations increase

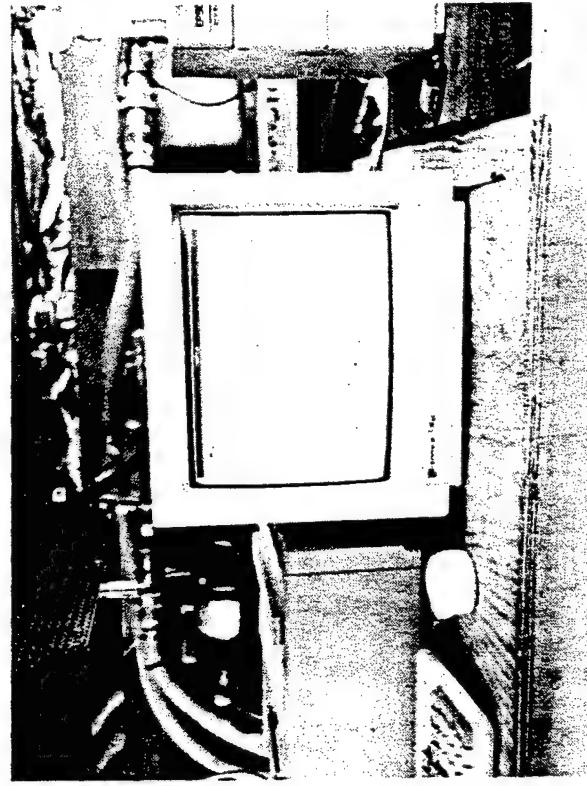
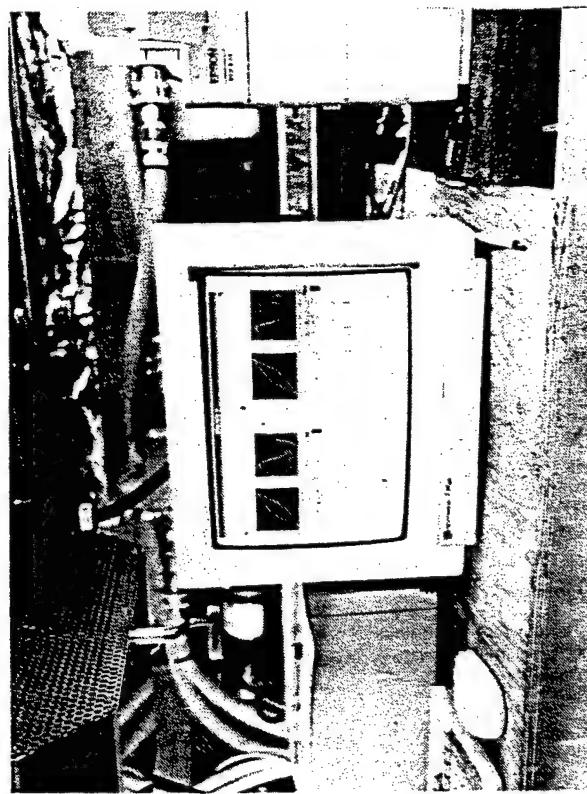


Assembling two 5-SCFM (142 lit/min) SDP processor units, each containing two 10-cell modules, in the equipment trailer at McClellan Air Force Base. The plasma power for each 5-SCFM unit is about 10 kW.



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Photos of the monitor which displays data from the computer-based data acquisition and control system for SDP remediation equipment. Photos taken during initial equipment shake-down tests at McClellan Air Force Base.



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Results from VOC tests at McClellan AFB (with prototype SDP equipment)

Sample #	Total [VOC] (ppmv)	Gas temperature (C)	Gas flow (SCFM)	Energy density (J/lit)	DRE (%)	Notes
1	542	32	10.0	4162	93.5	Wet
2	462	59	10.0	4193	88.1	Wet
3	328	58	10.0	4185	95.6	Wet
4	333	56	9.5	4416	90.0	Wet
5	363	50	10.4	4068	90.0	Wet
6	460	20	4.7	4494	97.7	Wet
7	477	38	5.4	4034	93.0	Wet
8	464	38	4.1	5075	92.5	Wet
9	532	55	4.1	5189	92.5	H ₂ /wet
10	629	50	5.1	4083	99.4	H ₂ /dry
11	698	18	3.7	5734	98.5	H ₂ /dry
12	459	24	2.9	7396	96.7	H ₂ /wet

Ideas for the future.

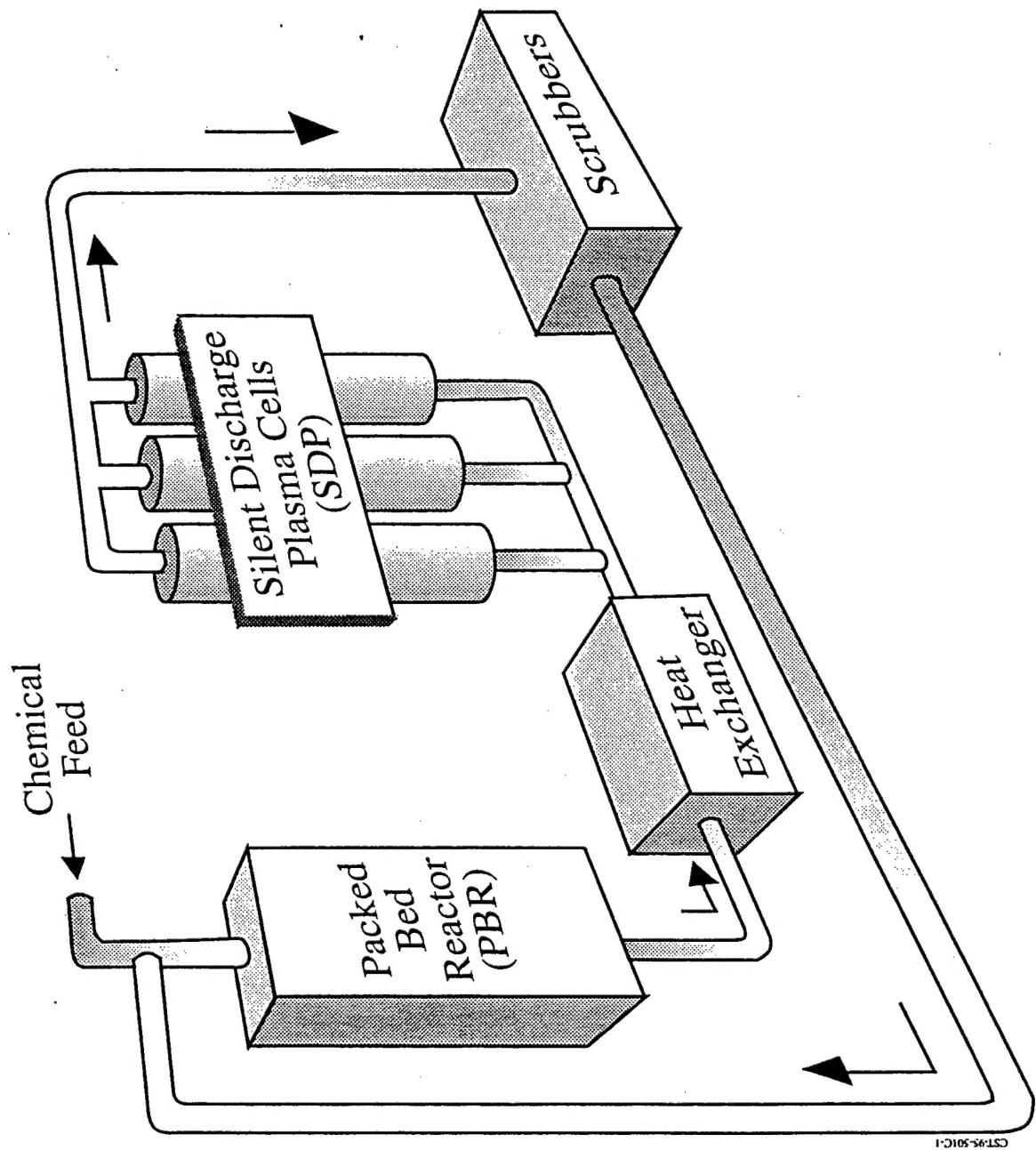
Finding market niches where NTP technology has significant advantages over conventional technologies in terms of:

- Increased efficiency and/or economics
- Increased selectivity or throughput
- Better control of final products and/or process
- Unique process streams

Potential examples:

- Regeneration of adsorbers (e.g., activated carbon) for environmental remediation and other air-pollution control applications
- Treatment of specialized radiochemical wastes

Two-Stage Thermal/Non-Thermal Hybrid Process for Hazardous Waste Destruction



Example: The two-stage process allows considerable flexibility in waste feeds and feed gases.

The operating conditions of the packed-bed reactor stage are flexible. We have demonstrated the ability to handle a wide variety of waste-feed surrogates, including those with high water content and different types of slurries.

In the non-thermal plasma stage, the yields of active species depend on the gas mixture.

A feed gas can be chosen to optimize the production of active species and the destruction of the waste.

Summary

- Most technical literature on treating air emissions with NTPs mainly presents phenomenological descriptions of reactor performance and, in most cases, does not provide a consistent way to compare and/or predict the scaling and optimization properties of different NTP systems.
- A simple way of comparing different types of NTP reactors, is based on the concept of: the plasma specific energy (electrical energy per unit volume deposited in the reactor active volume) required to remove a particular pollutant to a prescribed level in a defined exhaust-gas mixture and the associated yield (electrical energy cost per mass of pollutant removed).
- NTP is an emerging air-emissions control technology. Very few commercial systems exist. Also, for many emissions applications, the present forms of NTP technology are expected to be expensive (in terms of electrical power consumption) - and ancillary equipment (e.g., scrubbers) that may be necessary to handle treatment byproducts.
- Realizing the performance and economic shortcomings of stand-alone NTP reactors, the use of staged or hybrid systems to better match particular air-emissions control applications is proposed.
- Present trends for emerging NTP technologies are favorable. However, rigorous pilot-plant tests are required to provide further data and operating experience to more fully evaluate economic and performance projections.